Extraction of Am(III) from simulated Highly Active Raffinate solutions by soft-donor ligands dissolved in ionic liquid/molecular diluent mixtures

Peter Zsabka[a,b], Karen Van Hecke[a] , Simone Cagno[a], Marc Verwerft[a], Thomas Cardinaels[a,b], Koen Binnemans[b]

[a] *SCK•CEN, Institute for Nuclear Materials Science, Boeretang 200, 2400 Mol, Belgium*

[b] *KU Leuven, Department of Chemistry, Celestijnenlaan 200F, 3001 Leuven, Belgium*

* Email: pzsabka@sckcen.be

Long-lived isotopes of americium and curium (the so-called minor actinides, MA) are the main contributors to the heat production and high radiotoxicity of the nuclear waste after the decay of the shorter-lived fission products. These radionuclides make both reprocessing and the final disposal complicated and expensive. In the framework of the Belgian MYRRHA-project, a GenIV prototype accelerator-driven fast reactor will be built, in which these long-lived MA can be efficiently burnt.

If we consider a closed fuel cycle (and thus reprocessing), uranium and plutonium are separated from spent nuclear fuel via the so-called PUREX process, leaving a PUREX raffinate containing the fission products and minor actinides. In order to separate a small amount of trivalent actinide elements from the PUREX raffinate, several sophisticated processes have been elaborated (e.g. TALSPEAK, DIAMEX, SANEX and GANEX), all of which are based on the use of aliphatic diluents such as *n*-dodecane or kerosene. These diluents however all have common shortcomings, namely they are volatile, flammable and sensitive to radiation-induced degradation that reduces their process lifetime. By the application of non-volatile, non-flammable and radiation-resistant ionic liquid diluent for the minor actinide separation, secondary waste production can be reduced.

A fully incinerable solvent mixture of the quaternary ammonium ionic liquid [Aliquat-336][NO_3] and 1-octanol with soft-donor ligands were used for the liquid-liquid extraction of MA from nitric acid solutions. Molecular diluents reduce the viscosity of the ionic liquid; they not only improve extraction kinetics, but also facilitate phase disengagement. A high concentration of nitrate ions in the organic phase is ensured by the ionic liquid component. This is beneficial as the nitrate ions are effective quenchers for high energy radicals formed by the absorbed radiation. Studies with the 1-,10-phenanthroline-derived N-donor ligand $CyMe_4$BTPhen as extractant in [Aliquat-336][NO_3] diluent have shown that the extraction of MA is irreversible. Addition of a molecular diluent made the back-extraction feasible and significantly reduced the solvent viscosity.

Batch extractions were conducted using stable lanthanide isotopes and a simulated highly active raffinate (HAR) solution, spiked with ^{241}Am tracer in order to determine the distribution ratios, separation factors, and kinetics of the extraction process, and also the effect of loading on the phase behavior of the ionic liquids. The selective removal or selective masking of interfering elements present in the HAR (Zr, Pd, Rh, Ru) was also addressed.

Finally, speciation studies on the binary solvents were performed in order to gain a mechanistic insight in the change in complex structure upon addition of molecular diluents.